

CLAIMS

1. A method for producing an aromatic carbonate, comprising:

5 (1) performing a reaction between an organometal compound and carbon dioxide to obtain a reaction mixture containing a dialkyl carbonate formed by the reaction,

(2) separating said dialkyl carbonate from said  
10 reaction mixture to obtain a residual liquid, and

performing the following steps (3) and (4) in either order, or partially or wholly simultaneously:

(3) reacting said residual liquid with an alcohol to form at least one organometal compound and form water and removing said water from said organometal compound, and  
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(4) reacting said dialkyl carbonate separated in step (2) with an aromatic hydroxy compound to obtain an aromatic carbonate.

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2. The method according to claim 1, wherein said aromatic carbonate obtained in step (4) is at least one compound selected from the group consisting of an alkyl aryl carbonate and a diaryl carbonate.

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3. The method according to claim 1 or 2, wherein, in step (3), said organometal compound having said water removed therefrom is recycled to step (1).

5 4. The method according to any one of claims 1 to 3, wherein, in step (4), an alcohol which is generated together with said aromatic carbonate is recycled to step (3).

10 5. The method according to any one of claims 1 to 4, wherein a dialkyl carbonate recovered in step (4) is recycled to step (4).

15 6. The method according to any one of claims 1 to 5, wherein a cycle of steps (1) to (4) is repeated at least one time.

20 7. The method according to any one of claims 2 to 5, wherein said aromatic carbonate obtained in step (4) is an alkyl aryl carbonate and which, after step (4), further comprises the following step (5):

(5) subjecting said alkyl aryl carbonate to a disproportionation reaction to obtain a diaryl carbonate.

25 8. The method according to claim 7, wherein, in step

(5), a dialkyl carbonate which is generated together with said diaryl carbonate is recycled to step (4).

5        9.    The method according to claim 7 or 8, wherein a cycle of steps (1) to (5) is repeated at least one time.

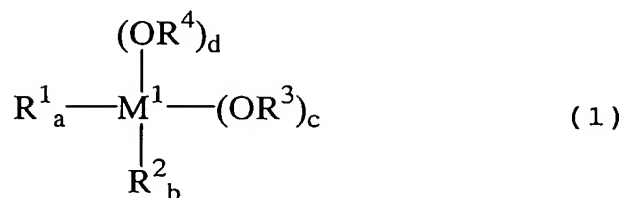
10       10.   The method according to any one of claims 1 to 9, wherein, in step (1), said organometal compound is used in an amount which is 1/200 to 1 time the stoichiometric amount relative to the amount of said carbon dioxide.

15       11.   The method according to any one of claims 1 to 10, wherein said reaction in step (1) is performed at 20 °C or higher.

20       12.   The method according to any one of claims 1 to 11, wherein said organometal compound used in step (1) is an organometal compound having a metal-oxygen-carbon linkage.

25       13.   The method according to claim 12, wherein said organometal compound having a metal-oxygen-carbon linkage comprises at least one compound selected from the group consisting of:

an organometal compound represented by the formula  
(1):



5            wherein:

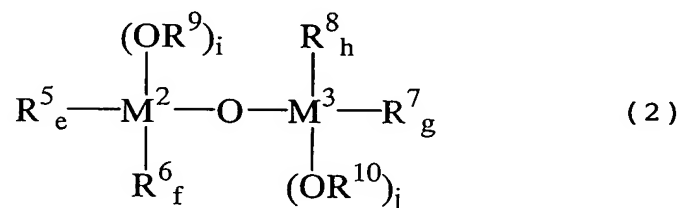
$\text{M}^1$  represents a metal atom selected from the group consisting of elements belonging to Groups 4 and 14 of the Periodic Table, exclusive of silicon;

10           each of  $\text{R}^1$  and  $\text{R}^2$  independently represents a straight chain or branched  $\text{C}_1$ - $\text{C}_{12}$  alkyl group, a  $\text{C}_5$ - $\text{C}_{12}$  cycloalkyl group, a straight chain or branched  $\text{C}_2$ - $\text{C}_{12}$  alkenyl group, a  $\text{C}_7$ - $\text{C}_{20}$  aralkyl group comprised of unsubstituted or substituted  
15            $\text{C}_6$ - $\text{C}_{19}$  aryl and alkyl selected from the group consisting of straight chain or branched  $\text{C}_1$ - $\text{C}_{14}$  alkyl and  $\text{C}_5$ - $\text{C}_{14}$  cycloalkyl, or an unsubstituted or substituted  $\text{C}_6$ - $\text{C}_{20}$  aryl group;

20           each of  $\text{R}^3$  and  $\text{R}^4$  independently represents a straight chain or branched  $\text{C}_1$ - $\text{C}_{12}$  alkyl group, a

C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, a straight chain or branched C<sub>2</sub>-C<sub>12</sub> alkenyl group, or a C<sub>7</sub>-C<sub>20</sub> aralkyl group comprised of unsubstituted or substituted C<sub>6</sub>-C<sub>19</sub> aryl and alkyl selected from the group consisting of straight chain or branched C<sub>1</sub>-C<sub>14</sub> alkyl and C<sub>5</sub>-C<sub>14</sub> cycloalkyl; and

each of a and b is an integer of from 0 to 2, a + b = 0 to 2, each of c and d is an integer of from 0 to 4, and a + b + c + d = 4; and an organometal compound represented by the formula (2):



wherein:

each of M<sup>2</sup> and M<sup>3</sup> independently represents a metal atom selected from the group consisting of elements belonging to Groups 4 and 14 of the Periodic Table, exclusive of silicon;

each of R<sup>5</sup>, R<sup>6</sup>, R<sup>7</sup> and R<sup>8</sup> independently represents a straight chain or branched C<sub>1</sub>-C<sub>12</sub> alkyl group, a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, a straight

chain or branched C<sub>2</sub>-C<sub>12</sub> alkenyl group, a C<sub>7</sub>-C<sub>20</sub> aralkyl group comprised of unsubstituted or substituted C<sub>6</sub>-C<sub>19</sub> aryl and alkyl selected from the group consisting of straight chain or branched C<sub>1</sub>-C<sub>14</sub> alkyl and C<sub>5</sub>-C<sub>14</sub> cycloalkyl, or an unsubstituted or substituted C<sub>6</sub>-C<sub>20</sub> aryl group;

each of R<sup>9</sup> and R<sup>10</sup> independently represents a straight chain or branched C<sub>1</sub>-C<sub>12</sub> alkyl group, a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, a straight chain or branched C<sub>2</sub>-C<sub>12</sub> alkenyl group, or a C<sub>7</sub>-C<sub>20</sub> aralkyl group comprised of unsubstituted or substituted C<sub>6</sub>-C<sub>19</sub> aryl and alkyl selected from the group consisting of straight chain or branched C<sub>1</sub>-C<sub>14</sub> alkyl and C<sub>5</sub>-C<sub>14</sub> cycloalkyl; and

each of e, f, g and h is an integer of from 0 to 2, e + f = 0 to 2, g + h = 0 to 2, each of i and j is an integer of from 1 to 3, e + f + i = 3, and g + h + j = 3.

14. The method according to any one of claims 1 to 13, wherein said separation of said dialkyl carbonate in step (2) is performed by at least one separation method selected from the group consisting of distillation, extraction and filtration.

15. The method according to claim 14, wherein said separation of said dialkyl carbonate in step (2) is performed by distillation.

5 16. The method according to claim 15, wherein said separation of said dialkyl carbonate in step (2) is performed by thin film distillation.

10 17. The method according to any one of claims 1 to 16, wherein said removal of said water in step (3) is performed by membrane separation.

18. The method according to claim 17, wherein said membrane separation is pervaporation.

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19. The method according to any one of claims 1 to 16, wherein said removal of said water in step (3) is performed by distillation.

20 20. The method according to any one of claims 1 to 19, wherein said alcohol used in step (3) is at least one alcohol selected from the group consisting of an alkyl alcohol having a straight chain or branched C<sub>1</sub>-C<sub>12</sub> alkyl group, a cycloalkyl alcohol having a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, an alkenyl alcohol having a straight chain

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or branched C<sub>2</sub>-C<sub>12</sub> alkenyl group, and an aralkyl alcohol having a C<sub>7</sub>-C<sub>20</sub> aralkyl group comprised of unsubstituted or substituted C<sub>6</sub>-C<sub>19</sub> aryl and alkyl selected from the group consisting of a straight chain or  
5 branched C<sub>1</sub>-C<sub>14</sub> alkyl and C<sub>5</sub>-C<sub>14</sub> cycloalkyl.

21. The method according to any one of claims 1 to 20, wherein the alcohol used in step (3) has a boiling point which is higher than the boiling point of water.

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22. The method according to claim 21, wherein the alcohol used in step (3) is at least one alcohol selected from the group consisting of 1-butanol, 2-methyl-1-propanol, an alkyl alcohol having a straight chain or  
15 branched C<sub>5</sub>-C<sub>12</sub> alkyl group, and an alkenyl alcohol having a straight chain or branched C<sub>4</sub>-C<sub>12</sub> alkenyl group.

23. The method according to claim 21 or 22, wherein  
20 the alcohol used in step (3) has a boiling point which is lower than that of said aromatic hydroxy compound used in step (4).

24. The method according to claim 13, wherein each of  
25 R<sup>3</sup> and R<sup>4</sup> in formula (1) and R<sup>9</sup> and R<sup>10</sup> in formula (2)



independently represents an n-butyl group, a 2-methylpropyl group, a straight chain or branched C<sub>5</sub>-C<sub>12</sub> alkyl group, or a straight chain or branched C<sub>4</sub>-C<sub>12</sub> alkenyl group.

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25. The method according to any one of claims 1 to 24, wherein, in step (1), said organometal compound is used in at least one form selected from the group consisting of a monomeric form, an oligomeric form, a polymeric  
10 form and an associated form.

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26. The method according to any one of claims 13, 24 and 25, wherein each of M<sup>1</sup> in formula (1) and M<sup>2</sup> and M<sup>3</sup> in formula (2) represents a tin atom.

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27. The method according to any one of claims 1 to 26, wherein said organometal compound used in step (1) is produced from an organotin oxide and an alcohol.

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28. The method according to any one of claims 1 to 27, wherein, the amount of said aromatic hydroxy compound used in step (4) is 0.1 to 10,000 times the stoichiometric amount relative to the amount of said dialkyl carbonate used in step (4).

29. The method according to any one of claims 1 to 28, wherein said reaction in step (4) is performed at a temperature in the range of from 50 to 350 °C.

5 30. The method according to any one of claims 1 to 29, wherein said reaction in step (4) is performed in the presence of a transesterification reaction catalyst.

31. The method according to any one of claims 7 to 30,  
10 wherein said reaction in step (5) is performed in the presence of a disproportionation reaction catalyst.

32. The method according to any one of claims 1 to 31,  
15 wherein said aromatic hydroxy compound is represented by the following formula (3):



wherein Ar is a C<sub>5</sub>-C<sub>30</sub> aromatic group.

20 33. The method according to claim 32, wherein said aromatic hydroxy compound represented by formula (3) is phenol.

34. The method according to any one of claims 1 to 33,  
25 wherein the total content of an aromatic hydroxy com-

pound and a carboxyl group-containing compound in said alcohol used in step (3) is 1,000 ppm or less.

5 35. An aromatic carbonate produced by the method of any one of claims 1 to 34.

36. A polycarbonate, an isocyanate or a polycarbonate diol produced using the aromatic carbonate of claim 35.

10 37. The polycarbonate, isocyanate or polycarbonate diol according to claim 36, wherein said aromatic carbonate is a diaryl carbonate.